Effect of Co doping on superconductivity and transport properties in $SmFe_{1-x}Co_xAsO$

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We investigate superconductivity and transport properties of Co doped $SmFe_{1-x}Co_xAsO$ system. The antiferromagnetic (AFM) spin-density wave (SDW) order is rapidly suppressed by Co doping, and superconductivity emerges as $x \geq 0.05$. $T_c{}^{mid}$ increases with increasing Co content, shows a maximum of 17.2 K at the optimally doping of $x \sim 0.10$. A phase diagram is derived based on the transport measurements and a dome-like T_c versus x curve is established. Meanwhile we found that the normal state thermopower might consist of two different contributions. One contribution increases gradually with increasing x, and the other contribution is abnormally enhanced in the superconducting window $0.05 \leq x \leq 0.20$, and shows a dome-like doping dependence. A close correlation between T_c and the abnormally enhanced term of thermopower is proposed.

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I. INTRODUCTION

Soon after the discovery of superconductivity at 26 K in $LaO_{1-x}F_xFeAs$[1], the substitution of La by other rare earth elements such as Ce[2], Pr[3], Sm[4,5], Nd[5], Gd[6,7], and Tb[8,9] has led to a family of 1111 phase high-T_c superconductors. The parent compounds of the new 1111 phase materials, LnFeAsO (Ln = La, Ce, Pr, Sm, Nd, Gd, and Tb etc.), have a quasi two-dimensional tetragonal structure, consisting of insulating Ln_2O_2 layers and conducting Fe_2As_2 layers. Similar to high-T_c cuprates, superconductivity occurs through electron (or hole) doping into an antiferromagentic (AFM) parent compound. In the high-T_c cuprates, superconductivity is induced by chemical doping in "charge reservoir" layers which are out of the superconducting CuO_2 planes. Meanwhile, the substitution of Cu with other $3d$ elements such as Ni and Zn in the CuO_2 planes severely destroys the superconductivity[10,11]. In contrast to high-T_c cuprates, it has been found that superconductivity can also be induced by partial substitution of Fe by other transition metal elements in the superconducting-active Fe_2As_2 layers. Sefat *et al.*[12] reported first the superconductivity in the Co doped $LaFe_{1-x}Co_xFeAs$. We also independently found that both Co doping[13] and Ni doping[14] can induce superconductivity in the LaOFeAs system. It is also found that the doping of non-magnetic impurities Zn^{2+} ions in the Fe_2As_2 conducting layers affects selectively the AFM order, and superconductivity remains almost unperturbed in $LaFe_{1-x}Zn_xAsO_{1-y}F_y$ system[15]. This implies that superconductivity is quite robust to the disorder in the conducting Fe_2As_2 layer, which might be taken for a significant difference between the high-T_c cuprates and iron-based arsenide superconductors.

Compared to the phase diagram of F-doped LaFeAsO[1,16,17], the phase diagram of Co-doped LaFeAsO system shows some significant differences[13]. Firstly, Co doping destroys the AFM SDW order more strongly. Secondly, the maximum T_c is significantly lowered in Co-doped system. Finally the optimal doping level is distinctly lower and the superconducting window is much narrower in Co doped $LaFe_{1-x}Co_xAsO$ system. The disorder effect caused by Co doping within (Fe/Co)As layers can not be ignored.

In this paper, we investigate in detail the Co doping effect on the superconductivity and transport properties of $SmFe_{1-x}Co_xAsO$, and an electronic phase diagram is derived. A dome-like Co doping (x) dependence of T_c is established. Furthermore, we have found that the normal state thermopower increases remarkably with Co doping in the "underdoped" region. A possible correlation between T_c and the enhanced thermopower in the superconducting window ($0.05 \leq x \leq 0.20$) is proposed.

II. EXPERIMENTAL

The polycrystalline samples of $SmFe_{1-x}Co_xAsO$ were prepared in vacuum by solid state reaction using SmAs, Sm_2O_3, Fe_2As, FeAs, and Co_3O_4 as starting materials. SmAs was pre-synthesized by reacting Sm slices and As powders at 1173 K for 24 hours in an evacuated quartz tube. FeAs and Fe_2As were obtained by reacting the mixture of stoichiometric element powders at 873 K for 10 hours, respectively. Co_3O_4 and La_2O_3 were dried in air at 773 K and 1173 K, respectively, for 24 hours before using. Then all powders of these intermediate materials were accurately weighed according to the stoichiometric ratio of $SmFe_{1-x}Co_xAsO$ (x=0, 0.01, 0.025, 0.05, 0.075, 0.1, 0.125, 0.15, 0.175, 0.2, 0.225, 0.25 and 0.3), thoroughly mixed in an agate mortar, and pressed into pellets under a pressure of 2000 kg/cm^2. All the processes were operated in a glove box filled with high-purity argon. Finally the pellets were sintered in an evacuated quartz tube at 1423 K for 40 hours and furnace-cooled to room temperature.

Powder X-ray diffraction (XRD) was performed at

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room temperature using a D/Max-rA diffractometer with Cu K_α radiation and a graphite monochromator. Lattice parameters were calculated by a least-square fit using at least 20 XRD peaks in the range of $20° \leq 2\theta \leq 80°$. The errors were estimated as three times of the standard deviations of the fit. The electrical resistivity was measured by four-terminal method. The temperature dependence of d.c. magnetization was measured on a Quantum Design magnetic property measurement system (MPMS-5) with an applied field of 10 Oe. The thermopower was measured by a steady-state technique, and the applied temperature gradient was less than 0.5 K/mm.

III. RESULTS AND DISCUSSION

Fig.1(a) shows the representative XRD patterns of the $SmFe_{1-x}Co_xAsO$ samples. The diffraction peaks of all the samples can be well indexed based on a tetragonal cell of ZrCuSiAs-type structure, which indicates that the samples are all pure phase. Fig.1(b) shows the variations of refined lattice parameters with Co content(x). Co doping causes the shrinkage of c-axis significantly, while the a-axis remains nearly unchanged. Thus the cell volume decreases monotonously with x, which is related to the smaller Co^{2+} ions (than Fe^{2+} ions). This fact indicates that Co is successfully doped into the lattice, according to Vegard's law. The shrinkage of c-axis suggests the strengthening of interlayer Coulomb attraction, implying the increase of density of negative charge in Fe_2As_2 layers by the Co doping. Similar variations of lattice constants were also observed in the Co doped $LaFe_{1-x}Co_xAsO$ in the previous report[13].

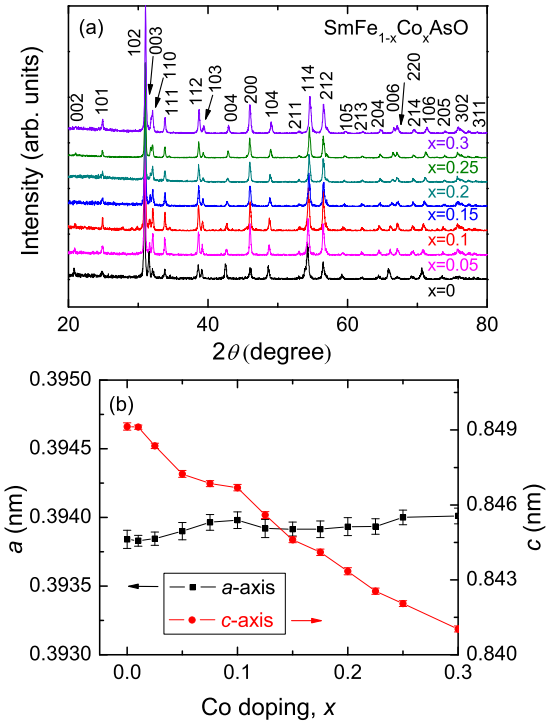

FIG. 1: (Color online) Structural characterization of $SmFe_{1-x}Co_xAsO$ samples. (a) Powder X-ray diffraction patterns of representative $SmFe_{1-x}Co_xAsO$ samples. (b) Lattice parameters as a function of Co content.

Fig.2 shows the temperature dependence of electrical resistivity (ρ) of $SmFe_{1-x}Co_xAsO$ samples in the temperature range from 3 K to 300 K. The inset shows an enlarged plot of ρ versus T for the low temperatures. For the undoped parent compound, a clear drop in the resistivity is observed below about 140 K just as in the case of LnOFeAs[1], which has been ascribed to a structural phase transition and antiferromagnetic spin-density-wave (SDW) transition[18,19]. This anomalous temperature T_{an}, which is defined as the peak position in the temperature dependence of the derivative of resistivity, decreases from 137 K for x =0, to 124 K and 93 K for $x = 0.01$ and 0.025, respectively. For $x = 0.05$, such an anomalous change in resistivity almost disappears, and only a tiny kink around 45 K can be distinguished. Within the doping range $0.05 \leq x \leq 0.20$, superconducting transition can be observed at low temperatures. Meanwhile, the resistivity anomaly disappears completely for $x > 0.05$. This means that the superconductivity occurs wherefrom the suppression of SDW order. Superconducting transition temperature T_c^{mid}, which is defined as the midpoint in the resistive transition, reaches a maximum of 17.2 K at the "optimally doped" level $x = 0.1$. This maximum of T_c^{mid} is larger than that of Co-doped $LaFe_{1-x}Co_xAsO$ system[13]. The volume fraction of magnetic shielding is over 60% for the "optimally" doped sample estimated according to its magnetic susceptibility (not shown here). Furthermore, the "superconducting window" is in the doping range $0.05 \leq x \leq 0.20$, which is also larger compared to the superconducting window ($0.025 \leq x \leq 0.125$) for $LaFe_{1-x}Co_xAsO$ system.

Another interesting feature is that there exists a resistivity minimum at T_{min} in the normal state in the underdoped and overdoped regimes. The resistivity changes from metallic into semiconductor-like as $T < T_{min}$, *i.e.*, there exists a crossover from metal into insulator as T decreases. However, such a resistivity upturn disappears in the doping regime $0.15 \leq x \leq 0.175$. We suggest that this upturn could be hidden in the strong superconducting fluctuations as T_c^{onset}, the onset point in the resistive transition, is quite high in this regime. Actually, such a crossover persists to $x > 0.20$ for Co doped $LaFe_{1-x}Co_xAsO$ system[13]. Meanwhile the room temperature resistivity shows a monotonous decrease with increasing x. In the region of large Co doping level ($x > 0.15$), the temperature dependence of resistivity follows a power law for temperature range $T > T_{min}$, *i.e.*, $\rho \propto T^n$. The index n is about 1.65 for $x = 0.25$. It is clear that the system becomes more metallic with increasing Co content, consistent with the itinerant charac-

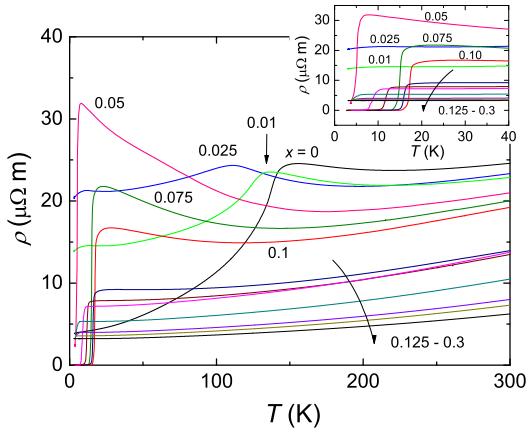

FIG. 2: (Color online) Temperature dependence of resistivity (ρ) for the $SmFe_{1-x}Co_xAsO$ samples. Inset: the enlarged plot of ρ verus T for low temperatures, showing the superconducting transitions.

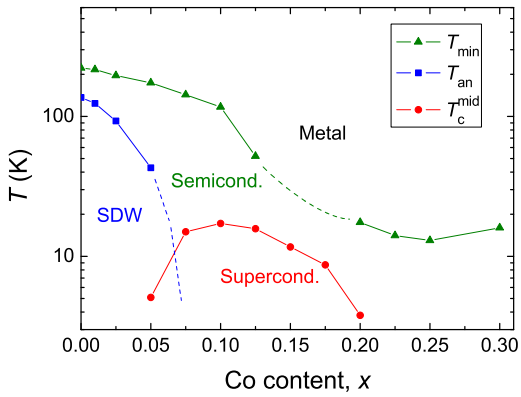

FIG. 3: (Color online) The electronic phase diagram for $SmFe_{1-x}Co_xAsO$. T_{min} separates the metallic and semiconducting regions in the normal state of the superconductors. Note that the vertical axis is in logarithmic scale.

ter of Fe $3d$ electrons in the iron-based oxy-arsenides revealed by the band structure calculations and theoretical analysis[16,20,21]. Furthermore, the theoretic calculation[22] reveals that total electron density-of-states (DOS) for $LaOMAs$ (M = Mn, Fe, Co and Ni) remains basically unchanged, except that Fermi level shifts toward the top of valence band with band filling (adding electrons) one by one from M = Mn, Fe, Co to Ni. According to this calculation, partial substitution of iron by cobalt is expected to add electrons into Fe_2As_2 layers, and thus the more metallic state is expected with increasing x.

As noted in the previous report[13], the possibility that oxygen deficiency itself might induce superconductivity in this system can be excluded. By high-pressure synthesis, superconductivity was indeed observed in oxygen-deficient $LnFeAsO_{1-\delta}$[23,24]. It has also been reported that superconductivity was induced by oxygen deficiency in Sr-doped LaFeAsO via annealing in vaccum.[25] We note that all the reported superconductors showed a *remarkable* decrease in a-axis as well as c-axis owing to the oxygen deficiency. In contrast, the present $SmFe_{1-x}Co_xAsO$ samples show no obvious change in the a-axis, suggesting no significant oxygen deficiency.

Based on above resistivity data, an electronic phase diagram for $SmFe_{1-x}Co_xAsO$ was thus established, as depicted in Fig. 3. The phase region of the SDW state is very narrow. 5% Co doping completely destroys the SDW order, and superconductivity emerges. In the superconducting window ($0.05 \leq x \leq 0.20$), a dome-like $T_c(x)$ curve is observed, similar to that of cuprate superconductors. Similar dome-like doping denpendence of T_c is also established for $LaFe_{1-x}Co_xAsO$[13]. However, the superconducting window of $SmFe_{1-x}Co_xAsO$ system is larger compared to that of $LaFe_{1-x}Co_xAsO$ system. Though the normal state shows metallic conduction at high temperatures, the upturn in resistivity is observed at low temperatures in a large doping region. For the higher Co-doping levels ($x \geq 0.20$), superconductivity no longer survives, but the resistivity becomes more metallic. Complete replacement of Fe by Co is possible, but whether SmCoAsO is an itinerant ferromagnetic metal like LaCoAsO[26] need to be clarified.

Fig. 4 shows the temperature dependence of normal state thermopower (S) for $SmFe_{1-x}Co_xAsO$ samples. All of the thermopowers are negative in the whole temperature range, which means that the electron-like charge carriers dominate. For the undoped parent compound, thermopower starts to increase abnormally around T_{an} at which the resistivity starts to decrease. Similar anomalous increase in the thermopower below T_{an} has been reported in the undoped parent compounds LaFeAsO[31] and TbFeAsO[9]. Such a remarkable change in the thermopower should be caused by the change in the electronic state when the system undergoes the structural phase transition and SDW transition. This anomaly is gradually suppressed with increasing Co doping, and disappears for $x > 0.05$, consistent with the resistivity data. For the superconducting samples, the profile of $S(T)$ curves is very similar to that of high-T_c cuprates except that it is negative for $SmFe_{1-x}Co_xAsO$ sytem. However, in contrast to high-T_c cuprates where the value of normal state thermopower decreases monotonously with increasing doping level[27,28], the absolute value of thermopower, $|S|$, increases quickly with Co doping, and the maximum in $|S|$ is about 80 μV/K for optimally doped level ($x = 0.1$). Such a large value of $|S|$ is very unusual in superconducting materials. However, the remarkable enhancement of $|S|$ has also been observed in F doped $LaFeAsO_{1-x}F_x$[29,30,31] and in Th doped $Tb_{1-x}Th_xFeAsO$[9]. It should be a universal feature for iron-based arsenide superconductors. It has also

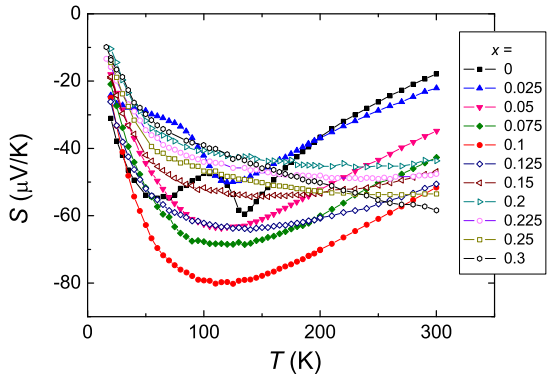

FIG. 4: (Color online) Temperature dependence of thermopower (S) for $SmFe_{1-x}Co_xAsO$ samples.

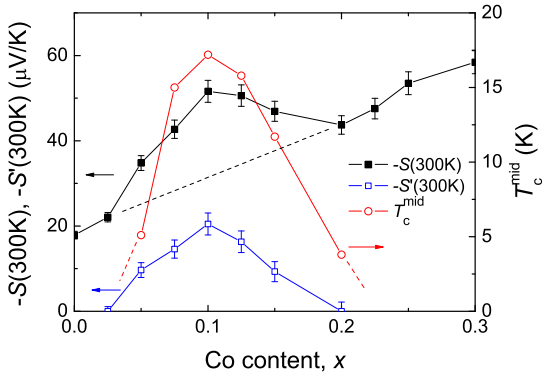

FIG. 5: (Color online) Doping dependence of room-temperature thermopower, $S(300)$, for $SmFe_{1-x}Co_xAsO$ samples. The superconducting transition temperature T_c^{mid} is also shown for comparison. The dashed line indicates the background term to the thermopower. $S'(300K)$ is the abnormally enhanced term, equal to $S(300K)$ subtracting the background normal term. See text for detail.

been proposed that the F-doped iron-oxypnictides can be promising thermoelectric materials in refrigeration applications around liquid nitrogen temperatures[29]. A rough estimate of $|S|$ according to the Mott expression gives a value of less than 10 μV/K for F doped LnFeAsO[30]. Whether the enhanced thermopower is associated with strong electron correlation, magnetic fluctuations, or specific electronic structure is an open issue.

It is well established that there is a universal doping (hole concentration) dependence of superconducting transition temperature, T_c, for high-T_c cuprates. Furthermore, it has been found that there exists a close correlation between the room temperature thermopower, $S(290K)$, and the hole concentration, p, and thus a universal correlation between T_c and $S(290K)$ is observed[27,28]. In order to explore the possible relationship between thermopower and superconducting transition temperature in this system, we also plot both $S(300K)$ and T_c^{mid} versus the doping level (x) for $SmFe_{1-x}Co_xAsO$ sytem. It becomes obvious that $S(300K)$ increases with x as T_c^{mid} does for $x < 0.1$, reaches a maximum at $x = 0.1$, and then gradually decreases with x in the overdoped region. For $x > 0.2$, superconductivity disappears and the thermopower starts to increase again. Actually it can be seen from Fig.5, that there seems to be two different contributions to the thermopower, *i.e.*, $S(300K) = S_0(300K) + S'(300K)$. The first term $S_0(300K)$ is the normal contribution (shown by the dashed line in the superconducting window), which increase gradually with increasing x. The other term $S'(300K)$ only appears in the superocnducting window (shown by the blue open symbols in Fig.5), which shows a dome-like doping dependence as T_c^{mid} does. We propose that there should be a close correlation between superconducting state and the anomalous term $S'(300K)$. It will be an interesting issue whether such a correlation between T_c and $S'(300K)$ is a universal feature for all the iron-based arsenide superconductors.

The anomalous contribution to the thermopower, represented by $|S'|(300K)$, is hard to understand in frame of conventional metal. We note that the thermopower of a cobaltate Na_xCoO_2 is remarkably enhanced due to the electronic spin entropy[32]. Thus we suggest that the anomalous thermopower term might have a magnetic origin. Careful studies on the d.c. magnetic susceptibility have found that the normal state magnetic susceptibility shows indeed a dome-like doping dependence in F-doped $LaFeAsO_{1-x}F_x$ system[33]. This susceptibility enhancement could be associated with spin fluctuations. Therefore it was proposed that the spin fluctuations may play an important role in the superconducting mechanism. However, the iron arsenide system has very different nature in electronic state compared to the sodium cobaltate system. In sodium cobaltate system, a strong electron correlation picture is necessary to describe electronic transport properties. The observation of suppression of thermopower by magnetic field suggested a large spin entropy term in thermopower. In contrast, the parent compounds LnFeAsO in the iron arsenide system are not Mott insulators, and band calculations[16,20,21,22] and transport property measurements have suggested that the 3d electrons in this system have itinerant nature. Therefore, the enhanced thermopower might not from the spin entropy although we argue that it might have a magnetic origin. How the spin fluctuations play an important role in the electronic transport need further studies. If both the enhanced thermopower and the enhanced susceptibility in the superconducting window have indeed the common origin, the magnetic fluctuations should also play an important role in the mechanism of superconductivity.

IV. CONCLUSION

In conclusion, superconductivity and normal state transport properties of Co doped $SmFe_{1-x}Co_xAsO$ system exhibit systematic variations with Co content. The SDW order is quickly suppressed by Co doping, and superconductivity emerges as $x \geq 0.05$. Meanwhile there is a crossover from metal to insulator in the normal state resistivity at low temperature. A phase diagram is derived based on the transport measurements and a dome-like T_c versus x curve is established. The maximum of T_c^{mid} is 17.2 K at the optimally doping level $x \sim 0.1$. Furthermore, thermopower increases with Co doping, also shows a maximum at $x \sim 0.10$, and then decreases slightly with decreasing T_c. After subtracting the background normal term, the anomalous term of the room temperature thermopower, $S'(300\text{K})$, also shows a dome-like doping dependence as $T_c^{mid}(x)$ does. A close correlation between T_c^{mid} and $S'(300\text{K})$ is proposed. This correlation may be associated with the mechanism of superconductivity.

Acknowledgments

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Note added. - At completion of this work we became aware of one paper by Y. Qi *et al.* which reported Co-doping induced superconductivity in $SmFe_{1-x}Co_xAsO$[34]. Only two Co doping concentrations were investigated, and T_c^{mid} of about 14.2 K was observed at $x = 0.10$ in their report. Their result is consistent with this paper.

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